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(6) DIFFUSION THEORY OF REACTION RATES, II.
ORNSTEIN – UHLENBECK APPROXIMATION.

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DIFFUSION THEORY OF REACTION RATES, II
ORNSTEIN--UHLENBECK APPROXIMATION

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February, 1978

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ABSTRACT

The diffusion theory of reaction rates is considered in the Ornstein--Uhlenbeck approximation. The rate constant is calculated by using a modified Kramers (eigenvalue) method and by using the expected time formulation. The transmission coefficient is calculated. It is shown how correlation functions can be calculated. In the last section, a model problem is considered.

DIFFUSION THEORY OF REACTION RATES, II
ORNSTEIN--UHLENBECK APPROXIMATION

SECTION 1.   INTRODUCTION

In the previous paper (1), denoted by I, we derived the diffusion theory of reaction rates (equations 2.16-2.29 of I). In the Einstein-Smoluchowski (ES) limit of large viscosity, we were able to calculate transmission coefficients, rate constants and lifetimes of activated species. The calculations were relatively straightforward because the ES limit eliminates one of the independent variables (the velocity) so that partial differential equations simplify to ordinary differential equation or to gradient systems. In this paper, we do not use the ES limit, but consider the full OU equations of I. Unlike in the analysis of I, we gain little by considering one dimensional problems in the OU approximation. The multidimensional OU equations are (see I):

$$\frac{\partial u}{\partial t} = \varepsilon \delta^{ij} \frac{\partial^2 u}{\partial v^i \partial v^j} + F^i(x)\frac{\partial u}{\partial v^i} + v^i \frac{\partial u}{\partial x^i} - v^i \frac{\partial u}{\partial v^i} \tag{1.1}$$

$$\frac{\partial \rho}{\partial t} = \varepsilon \delta^{ij} \frac{\partial^2 u}{\partial v^i \partial v^j} - v^i \frac{\partial \rho}{\partial x^i} - \frac{\partial}{\partial v^i} \left((F^i - v^i)\rho \right) \tag{1.2}$$

$$-u(x,v,\infty) = \varepsilon \delta^{ij} \frac{\partial^2 \bar{t}}{\partial v^i \partial v^j} + F^i \frac{\partial \bar{t}}{\partial v^i} + v^i \frac{\partial \bar{t}}{\partial x^i} - v^i \frac{\partial \bar{t}}{\partial v^i} \tag{1.3}$$

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In (1.1-1.3), repeated indices are summed from 1 to n. Namely (x,v) is a point in the $2n$ dimensional phase space. The force $F(x)$ is derived from a potential $F^i(x) = -\partial V/\partial x^i$; $V(x)$ is assumed to be known.

In the above equations

$$F(x) = -\nabla V(x) \tag{1.3a}$$

where $V(x)$ is the potential surface sketched in Fig. 1. The function $u(x,v,t)$ is defined by

$$u(x,v,t) = E\left\{ f\left(\tilde{x}(t), \tilde{v}(t)\right) \mid \tilde{x}(0) = x, \tilde{v}(0) = v\right\} \tag{1.4}$$

where $f(z,z_1)$ is an integrable function. By appropriate choices of f, we can construct the moments of $(\tilde{x},\tilde{v})$. The density for $(\tilde{x}(t), \tilde{v}(t))$ is $\rho(x,v,t)$:

$$\rho(x,v,t)dxdv = Pr\left\{\tilde{x}(t) \in (x, x + dx), \tilde{v}(t) \in (v, v + dv)\right\} \tag{1.5}$$

Finally $\overline{t}(x,v)$ is an expected time

$$\overline{t}(x,v) = \int_0^\infty t \ \frac{\partial u}{\partial t} (x,v,t)dt \tag{1.6}$$

In this paper, will will use equations (1.1-1.3) to calculate reaction rates and transmission coefficients. Our methods use the asymptotic techniques developed in (2) and (3). Since these
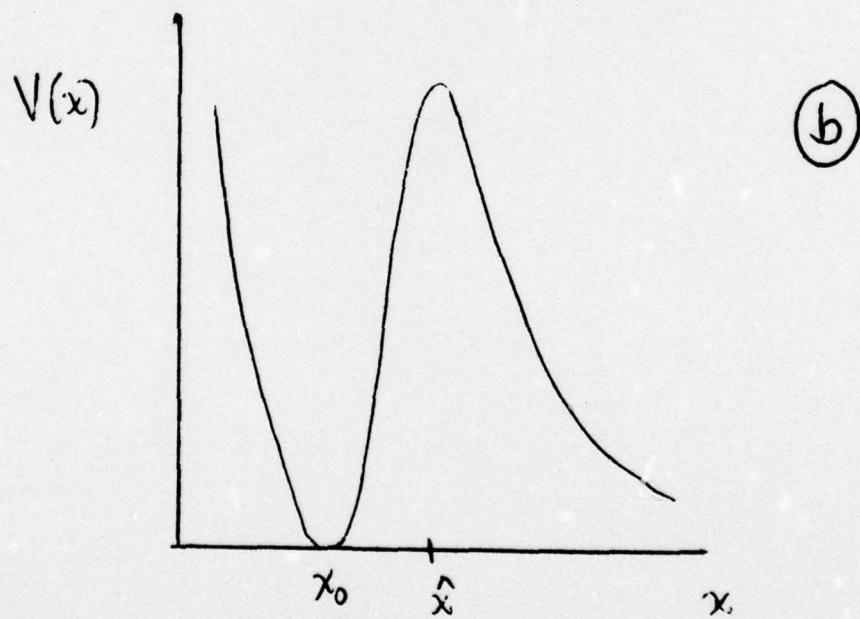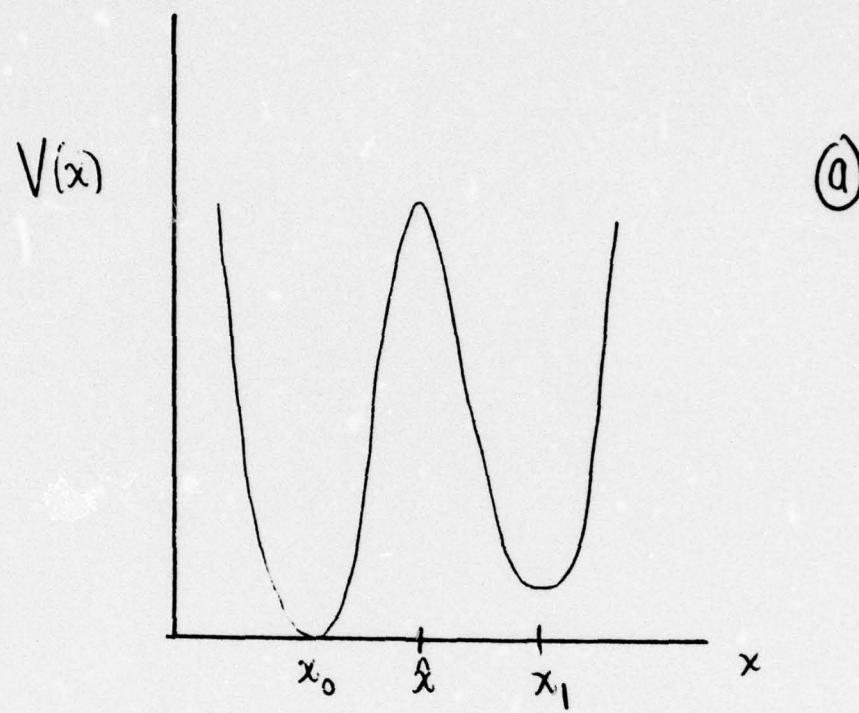
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FIG. 1: A CROSS SECTION OF THE MOLECULAR POTENTIAL V(x).
A) DOUBLE MINIMUM POTENTIAL SURFACE,
B) LENARD JONES POTENTIAL.

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techniques are quite involved, we will treat the problems in an order slightly different from that of I.

In addition to more complicated techniques, the formulation of the problems is more delicate. To illustrate some aspects of the delicacy, we will consider the phase plane in both OU and ES cases. (i.e. the special case $x \in R$). The phase plane corresponds to the deterministic equations obtained by averaging the equations (I. 2.17, 18,20.) These give

$$\frac{dx}{dt} = v \qquad \frac{dv}{dt} = F(x) - v \qquad (OU) \tag{1.7}$$

$$\frac{dx}{dt} = F(x) \qquad (ES) \tag{1.8}$$

The ES "phase plane" is just a line, sketched in Fig. 2. In this case, formulating problems in terms of rates to cross $\hat{x}$, or times to reach $\hat{x}$ is perfectly sensible. The OU phase plane is partially sketched in Fig. 3 (in §6 we give more detail). There is a trajectory S that plays the role of $\hat{x}$ in the ES case: S separates domains of attraction of $(x_0, 0)$ and $(x_1, 0)$ (double minimum case) or $(\infty, 0)$ (Lennard-Jones case) (see I). This trajectery is the deterministic separatrix. In the following, we shall use the separatrix quite a bit.
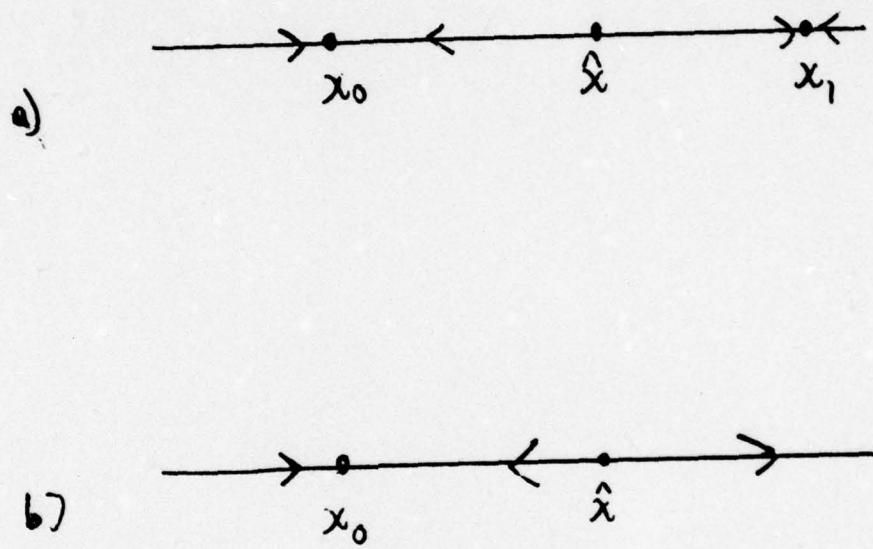
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FIG. 2:   THE ES PHASE SPACE.
          A) DOUBLE MINIMUM POTENTIAL SURFACE.
          B) LENARD JONES POTENTIAL SURFACE.

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For conceptual ease we connect the phase plane with the phase space, by assuming that the potential function $V(x)$ is quasi-separable. For example, if

$$V(x) = \tilde{V}(x^2, \ldots x^n) - \frac{1}{3}(x^1)^3 + \alpha x^1, \tag{1.9}$$

then the (x^1, v^1) phase plane will be similar to the one sketched in Fig. 3. The full phase space will be the direct product of the (x^1, v^1) phase plane and the $2n - 2$ dimensional $(x^2, \ldots x^n, v^2, \ldots v^n)$ phase space. Hence, we are assuming that the reaction coordinate is clearly defined (see, e.g. (4)).

In §2, we construct the reaction rate constant by calculating the lowest eigenvalue of the forward equation (1.2). We assume that the potential is of the form (1.9) and that the reaction proceeds along a path in which $(x^2, \ldots x^n)$ have equilibrium values. This assumption is made for notational convenience. In §3, we construct the reaction rate constant, under the same assumption about the potential, using the expected time formulation. In §4 we calculate the transmission coefficient. In §5, we illustrate how correlation functions, and thus spectra, can be found by using the "ray method". Finally, in §6, we consider a model problem that illustrates some of the ideas in §2-5.
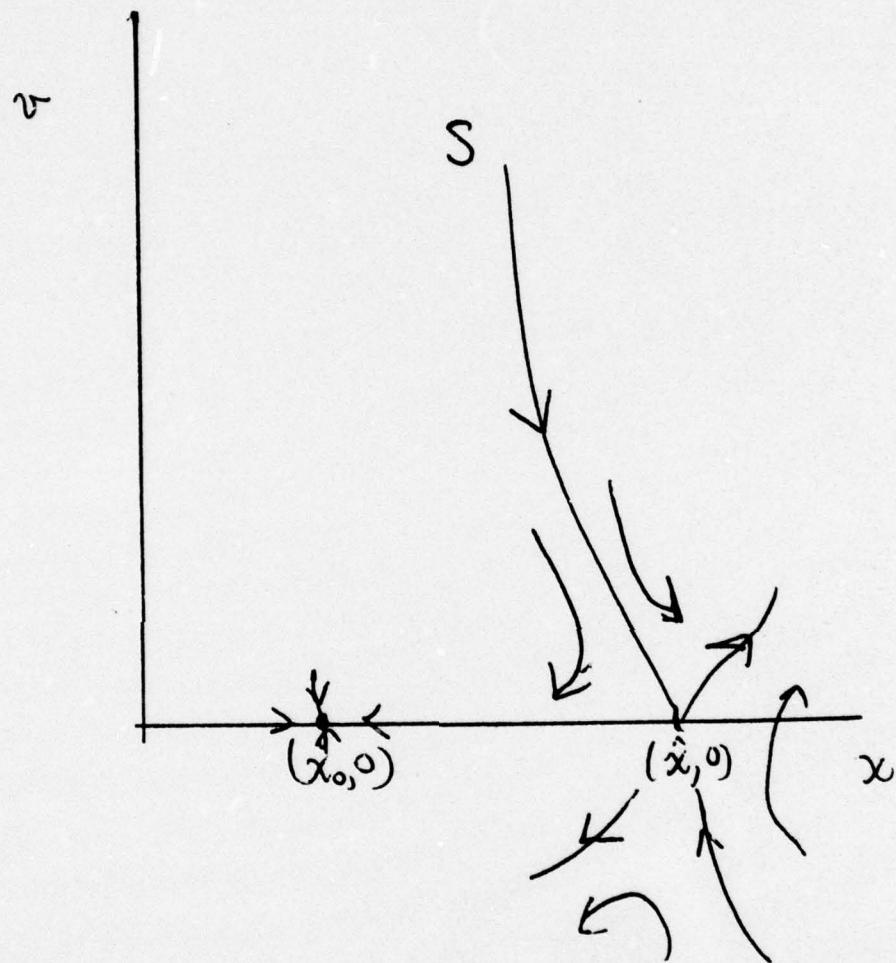
FIG. 3: THE OU PHASE SPACE. S IS THE DETERMINISTIC SEPARATRIX, $(\hat{x},0)$ IS A CLASSICAL SADDLE POINT.

SECTION 2

REACTION RATE CONSTANT BY MODIFIED KRAMERS METHOD

In this section, we use the Miller (5) -- Ludwig (2) theory to compute the rate at which particles pass from the well at x_0 (Fig. 1) to the barrier at $\hat{x}$. We seek a solution of (1.2) in the form

$$\hat{p}(x,v,t) = \sum_n \sigma_n(x,v) e^{-\lambda_n t} \tag{2.1}$$

In §2-6, we make the following assumptions about the reaction. They are made to ease the conceptualization of the ideas presented here and can easily be relaxed. First we assume that $V(x)$ has the form indicated in (1.9). Next, we assume that $(x^2, \ldots x^n, v^2, \ldots v^n)$ have relaxed to their equilibrium values in a time t_{rel} much smaller than the first passage time t_{fp} considered below. Thus, we reduce (1.1-1.3) to equations involving only x^1 and v^1, which we denote by (x,v). This type of assumption was used by Slater (6) and justified by Rice (7). In I we showed that the lowest eigenvalue, λ_0, is asymptotic to the rate at which molecules pass from x_0 to x_1. We are thus lead to consider the problem

$$-\lambda\sigma = \varepsilon \frac{\partial^2 \sigma}{\partial v^2} - v\frac{\partial \sigma}{\partial x} - \frac{\partial}{\partial v}\left(\left(F(x) - v\right)\sigma\right) \tag{2.2}$$

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$$\sigma(x, v) = 0$$

for $(x,v) \in S$ (2.3)

$$\lim_{v^2 \to \infty} \sigma(x,v) = 0$$

We now consider the semi-infinite domain in (x,v) space D consisting of all points to the left of $S = \partial D$. We integrate (2.2) over D and obtain

$$-\lambda = \frac{\iint_D \varepsilon \frac{\partial^2 \sigma}{\partial v^2} - v \frac{\partial \sigma}{\partial x} - \frac{\partial}{\partial v}\left(\left(F(x) - v\right)\sigma\right) dx dv}{\iint_D \sigma(x,v) dx dv} \qquad (2.4)$$

using Gauss' theorem, we obtain:

$$\lambda = \frac{\varepsilon \int_S \frac{v(t) \partial \sigma}{\partial v(t)} dt}{\iint_D \sigma(x,v) dx dv} \qquad (2.5)$$

In (2.6), t is the tangential coordinate along S and $x(t)$, $v(t)$ satisfy

$$\frac{dx}{dt} = v \qquad \frac{dv}{dt} = F(x) - v \qquad (2.6)$$

with $x(\infty) = \hat{x}$, $v(\infty) = 0$.

Following the argument in I, we now set

$$\sigma(x,v) = \exp\left[-\frac{1}{\varepsilon}\left(\frac{v^2}{2} + V(x)\right)\right] + O(\varepsilon) \tag{2.8}$$

Then using Laplace's method on the integrals in the numerator and denominator in (2.6), we obtain

$$\lambda \sim \frac{2}{\pi}\sqrt{V''(x_0)\,|V''(\hat{x})|}\ e^{-V(\hat{x})/\varepsilon}\,e^{V(x_0)/\varepsilon}, \tag{2.9}$$

the result (I, 5.16). It is interesting that the results (2.9) and (I, 5.16) agree exactly. They agree for the following reason. In the use of the above asymptotic method, we implicitly assume a rapid relaxation of the velocity, so that the particle crosses at $\hat{x}$ with v infintesimally small. Consequently, we make an implicit ES assumption, leading to (2.9).

SECTION 3

REACTION RATE CONSTANT BY EXPECTED TIME

The analysis in the previous section implicitly assumes that the barrier $V(\hat{x}) - V(x_0)$ is large, a point first noted by Kramers. (8) For many reactions, e.g. catalyzations, this is not the case. Hence, the procedure used in section 2 breaks down.

An alternative calculation of the rate constant uses the expected time formulation (see I, §5.2). We consider

$$-1 = \varepsilon \frac{\partial^2 \bar{t}}{\partial v^2} + F(x) \frac{\partial \bar{t}}{\partial v} + v \frac{\partial \bar{t}}{\partial x} - v \frac{\partial \bar{t}}{\partial v} \qquad (3.1)$$

with boundary conditions

$$\bar{t}(x,v) = 0 \qquad \lim_{v^2 \to \infty} \bar{t}(x,v) < \infty$$
$$(x,v) \in S$$

$$\lim_{x \to -\infty} \bar{t}(x,v) < \infty \qquad (3.2)$$

As defined in (3.2) $\bar{t}(x,v)$ is the expected time to reach S conditioned on $\tilde{x}(0) = x$, $\tilde{v}(0) = v$.

Let $\rho_0(x,0)$ be the initial distribution of molecules; it will be concentrated at $(x_0,0)$. We define the rate constant by

$$k = 1/<\bar{t}(x,v)>, \qquad (3.3)$$

where

$$\langle \overline{t}(x,v) \rangle = \iint_D \rho_0(x,v) \overline{t}(x,v) dx dv. \tag{3.4}$$

In (3.4), D is the domain consisting of all points to the left of the deterministic separatrix S.

To construct $t(x,v)$, we seek a solution of the form (3)

$$\overline{t}(x,v) = g(x,v,\varepsilon)B\ (\psi(x,v)/\varepsilon^{1/3}, \alpha/\varepsilon^{2/3}, 1/\varepsilon^{1/3}, \gamma)$$

$$+ \varepsilon^{2/3}h(x,v,\varepsilon)B'(\psi(x,v)/\varepsilon^{1/3}, \alpha/\varepsilon^{2/3}, 1/\varepsilon^{1/3}, \gamma)$$

$$+ k(x,v,\varepsilon) \tag{3.5}$$

In (3.5) $\psi(x,v)$, $g(x,v)$, $h(x,v)$, $k(x,v,\varepsilon)$, α and γ are to be determined. We assume that they have asymptotic expansions (3)

$$g(x,v,\varepsilon) = \sum g^n(x,v)\varepsilon^n \qquad h(x,v,\varepsilon) = \sum h^n(x,v)\varepsilon^n$$

$$k(x,v,\varepsilon) = \sum k^n(x,v)\varepsilon^n \qquad \alpha = \sum \alpha_n \varepsilon^n \tag{3.6}$$

$$\gamma = \sum \gamma_n \varepsilon^n$$

The function $B(z, \beta, 1, \gamma)$ satifies

$$\frac{d^2 B}{dz^2} = -(z^2 - \beta)\frac{dB}{dz} - 1 + \gamma z \tag{3.7}$$

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We shall, in fact, only consider the first term here. Higher order terms are treated in an analogous fashion. When derivatives are evaluated, (3.7) is used to replace B'' by $\left(\dfrac{\psi^2 - \alpha}{\epsilon^{2/3}}\right) B' - 1/\epsilon^{1/3}$ $+ \gamma\psi/\epsilon^{1/3}$. After derivatives are evaluated and substituted into (3.1), terms are collected according to powers of ϵ. The leading term vanishes if the following equations are satisfied:

$$O(\epsilon^{-1/3}B'): \quad \left(F(x) - v\right)\psi_v + v\psi_x - \psi_v^2(\psi^2 - \alpha_0) = 0 \qquad (3.8)$$

$$O(\epsilon^0 B): \quad \left(F(x) - v\right)g_v^0 + vg_x^0 = 0 \qquad (3.9)$$

$$O(\epsilon^0): \quad \left(F(x) - v\right)k_v^0 + k_x^0 + \psi_v^2 g^0(-1 + \gamma_0\psi) = -1 \qquad (3.10)$$

$$O(\epsilon^{2/3}B'): \quad \left(F(x) - v\right)h_v^0 + vh_x^0 - h^0\left(\psi_v(\psi^2 - \alpha_0)\right)_v \qquad (3.11)$$

$$-(\psi^2 - \alpha_0)h_v^0\psi_v - \alpha_1\psi_v^2\left(g^0 - (\psi^2 - \alpha_0)h^0\right) + g^0\psi_v v = 0$$

We first consider (3.8). In I, we showed that the classical (deterministic) equations for x, v are

$$\frac{dx}{dt} = v \qquad \frac{dv}{dt} = F(x) - v \qquad (3.12)$$

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These correspond to a set of "deterministic" trajectories in the (x,v) phase plane; parametrized by $x(0) = \bar{x}$ and $v(0) = \bar{v}$. The points $(x_0,0)$ and $(\hat{x},0)$ are, respectively, a metastable and unstable equilibrium of the system. The system is not a Hamiltonian system, due to the $-v$ term in the second equation in (3.12). Also, we are interested in an explicitly non-equilibrium phenomenon. Consequently, we require $\psi_x \neq 0$, $\psi_v \neq 0$ at either of the rest points. At these points, (3.8) becomes

$$\psi_v^2 (\psi^2 - \alpha_0) = 0 \tag{3.13}$$

We set (3)

$$\psi(x_0,0) = -\sqrt{\alpha_0} \qquad \psi(\hat{x},0) = \sqrt{\alpha_0} \tag{3.14}$$

The derivatives ψ_x, ψ_v at $(x_0,0)$ and $(\hat{x},0)$ can be determined by differentiating (3.8) with respect to x or v and evaluating the result at the rest points. Once these are known, the parameter α_0 can be calculated by the method of characteristics. Once α_0 is known $\psi(x,v)$ can be completely determined by the method of characteristics (3,9).

An argument using Hamilton-Jacobi theory (3) shows that $\psi = \sqrt{\alpha_0}$ on S. Hence the separatrix is a contour of ψ. If we set

$$B\left(\sqrt{\alpha_0}/\epsilon^{1/3}, \alpha/\epsilon^{2/3}, 1/\epsilon^{1/3}, \gamma\right) = B\left(\sqrt{\alpha_0}/\epsilon^{1/3}, \alpha/\epsilon^{2/3}, 1/\epsilon^{1/3}, \gamma\right) = 0 \tag{3.15}$$

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then the first two terms on (3.5) vanish identically on S.

Equation (3.9) incidates that g^0 is constant on trajectories. At $(x_0, 0)$ and $(\hat{x}, 0)$, equation (3.10) becomes

$$\psi_v^2(x_0, 0) g^0 (-1 - \gamma_0 \sqrt{\alpha_0}) = -1 \tag{3.16}$$

$$\psi_v^2(\hat{x}, 0) g^0 (-1 + \gamma_0 \sqrt{\alpha_0}) = -1 \tag{3.17}$$

These are two equations for the constants g^0 and γ_0. Next, we set $k_0(x, v) \equiv 0$ on S and solve (3.10) by the method of characteristics (9). With the above choice of initial data, $\bar{t}(x, v)$ vanishes identically on S.

On S, equation (3.11) becomes

$$\frac{dh^0}{dt} - 2h^0 \sqrt{\alpha_0} \quad \psi_v^2 = g^0 \psi_{vv} + \alpha_1 \psi_v^2 g^0 \tag{3.18}$$

At $(\hat{x}, 0)$ we obtain

$$h^0(\hat{x}, 0) = \frac{-(g^0 \psi_{vv} + \alpha_1 \psi^2 g^0)}{-2\psi_v^2 \sqrt{\alpha_0}} \quad . \tag{3.19}$$

Then h^0 can be determined on S, and in the entire phase plane by the method of characteristics. The parameter α_1 is determined in a manner analogous to the determination of α_0.

We have thus given a complete characterization of the first term:

$$\bar{t}(x,v) = g^0 B + \varepsilon^{2/3} h^0 B' + k^0 + 0(\varepsilon) \qquad (3.20)$$

With this construction, we can obtain the rate at which particles reach S from (x,v), as given by (3.3).

SECTION 4. TRANSMISSION COEFFICIENT

In this section, we give an asymptotic technique for the calculation of the transmission coefficient. As before, rather than using $(\hat{x},0)$ as a reference point, we use the separatrix S (Fig. 4). We surround S by a tube (S_1,S_2). Let

$$u(x) = \Pr\left\{\left(\tilde{x}(t),\tilde{v}(t)\right) \text{ crosses } S_2 \text{ before } S_1 \Big| \tilde{x}(0)\right.$$

$$\left. = x, \ \tilde{v}(0) = v\right\} \tag{4.1}$$

If S_1, S_2 are sufficiently far apart, then it is clear that $u(x,v)$ represents the probability that a molecule with initial reaction coordinate x and reaction coordinate velocity v becomes a product rather than returning to a reactant. The function $u(x,v)$ will satisfy

$$0 = \varepsilon\frac{\partial^2 u}{\partial v^2} + v\frac{\partial u}{\partial x} + \left(F(x) - v\right)\frac{\partial u}{\partial v} \tag{4.2}$$

with boundary conditions

$$u(x,v) = 0 \qquad (x,v) \in S_1 \tag{4.3}$$

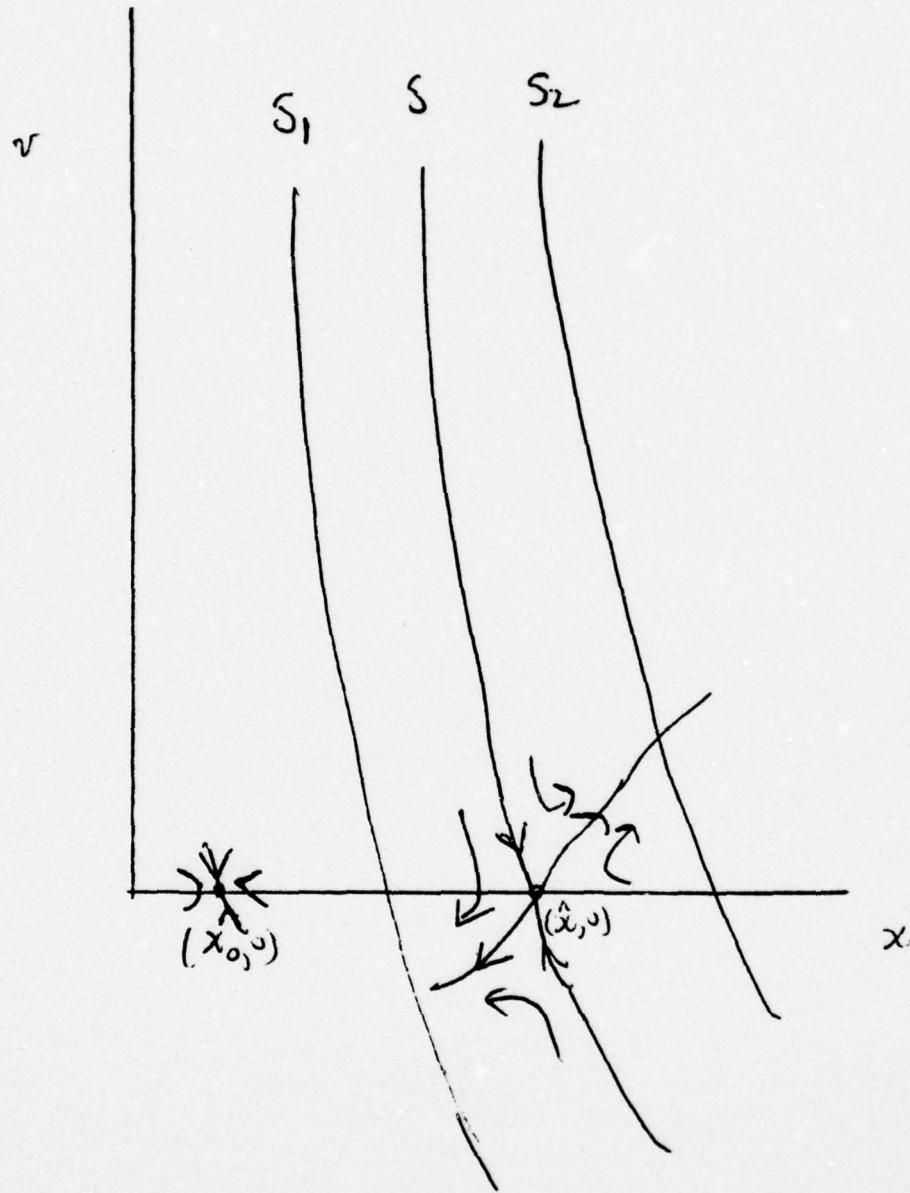$$u(x,v) = 1 \qquad (x,v) \in S_2 \tag{4.4}$$

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FIG. 4: BOUNDARIES FOR THE CALCULATION OF THE TRANSMISSION COEFFICIENT. THE TRANSMISSION COEFFICIENT IS THE PROBABILITY THAT A PARTICLE STARTING IN (S_1, S_2) EXITS THROUGH S_2.

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A formal asymptotic solution of (4.2) is (3)

$$u(x,v) = g(x,v,\varepsilon)E\left(\psi(x,v)/\sqrt{\varepsilon}\right) + \varepsilon^{1/2} h(x,v,\varepsilon)E'\left(\psi(x,v)/\sqrt{\varepsilon}\right)$$

(4.5)

with $\psi(x,v), g(x,v,\varepsilon)$ and $h(x,v,\varepsilon)$ to be determined. We assume that g and h have asymptotic expansions

$$g(x,v,\varepsilon) = \sum \varepsilon^n g^n(x,v)$$ (4.6)

$$h(x,v,\varepsilon) = \sum \varepsilon^n g^n(x,v)$$ (4.7)

In (4.5), $E(z)$ is the error function:

$$\frac{d^2E}{dz^2} = -z\frac{dE}{dz} \qquad z_0 \leq z \leq z_1$$ (4.8)

Following a procedure analogous to the one in the preceding section, we obtain equations for ψ, g^0 and h^0. (Higher order terms are treated treated in an analogous fashion).. These equations are:

$$v\psi_x + \left(F(x) - v\right)\psi_v - \psi\psi_v^2 = 0$$ (4.9)

$$vg^0{}_x + \left(F(x) - v\right)g_v^0 = 0$$ (4.10)

$$vh_x^0 + \left(F(x) - v\right)h_v^0 + g_v^0\psi_v - h_v^0\psi_v\psi$$ (4.11)

$$+ g^0\psi_v v - h^0\left((\psi\psi_v)v\right) = 0$$

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We require $\psi_x \neq 0$, $\psi_v \neq 0$ at $(\hat{x}, 0)$. Then (4.9) indicates that $\psi(\hat{x}, 0) = 0$. An argument using Hamilton-Jacobi theory shows that $\psi \equiv 0$ on S. When (4.9) is differentiated with respect to x,v and evaluated on S, we obtain

$$\frac{d}{dt} \psi_x + F_x(x)\psi_v - \psi_x \psi_v^2 = 0 \tag{4.12}$$

$$\frac{d}{dt} \psi_v + \psi_x - \psi_v - \psi_v^3 = 0 \tag{4.13}$$

In obtaining (4.12, 13) we used

$$\frac{d}{dt} \psi_k = v\psi_{kx} + \left(F(x) - v\right)\psi_{kv} \tag{4.14}$$

In light of (4.14), at $(\hat{x}, 0)$ (4.12, 13) become

$$F_x(x)\psi_v - \psi_x \psi_v^2 = 0 \tag{4.15}$$

$$\psi_x - \psi_v - \psi_v^3 = 0 \tag{4.16}$$

We denote the solution of these equations by $\overline{\psi}_x, \overline{\psi}_v$. Then (4.12, 13) are solved, subject to

$$\psi_x(\infty) = \overline{\psi}_x \qquad \psi_v(\infty) = \overline{\psi}_v \tag{4.17}$$

Thus, ψ_x and ψ_v are known along S. Consequently, $\psi(x, v)$ is known in the entire plane since (4.9) can be solved by the method of characteristics, with initial data on S (3,9).

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Let ψ_I denote the minimum value of ψ on S_1 and ψ_{II} the maximum value of ψ on S_2. Equation (4.10) indicates that g^0 is constant on trajectories. Following (3), we set z_0 in (4.8) equal to $\psi_I/\sqrt{\epsilon}$ and

$$g_0 = \left[E(\psi_{II}/\sqrt{\epsilon}) \right]^{-1} \tag{4.18}$$

If S_1 and S_2 happen to be level curves of ψ, then the choice (4.18) insures $u(x,v) \equiv 0$ on S_1 and $u(x,v) \equiv 1$ on S_2. Otherwise, u is exponentially small on S_1 and $1 - u$ is exponentially small on S_2.

Finally, we consider the equation (4.11) for h^0. On S, it becomes

$$\frac{dh^0}{dt} - h^0 \psi_v^2 = -g^0 \psi_{vv} \tag{4.19}$$

At $(\hat{x},0)$ equation (4.19) gives

$$h^0 = g^0 \overline{\psi}_{vv}/\overline{\psi}_v \equiv h^0(\infty) \tag{4.20}$$

where $\overline{\psi}_{vv} = \psi_{vv}(\hat{x},0)$. Then, on S, h^0 is given by

$$h^0(t) = \frac{\int_t^\infty g^0 \psi_{vv}(s) \exp\left[-\int^s \psi_v(t')^2 dt'\right] ds + h^0(\infty)}{\exp\left[-\int^t \psi_v(t')^2 dt'\right]} \tag{4.21}$$

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Once $h^0(t)$ is known on S, it can be calculated in the plane by the method of characteristics, as described in the previous section.

SECTION 5

CORRELATION FUNCTIONS AND SPECTRA

We now show how to calculate correlation functions and their Fourier transforms. We define

$$\phi_{xx}(\tau) = E\left\{\tilde{x}(t + \tau)\tilde{x}(t)\right\} \tag{5.1}$$

$$\phi_{xv}(\tau) = E\left\{\tilde{x}(t + \tau)\tilde{v}(t)\right\} = E\left\{\tilde{x}(t)\tilde{v}(t + \tau)\right\} \tag{5.2}$$

$$\phi_{vv}(\tau) = E\left\{\tilde{v}(t + \tau)\tilde{v}(t)\right\} \tag{5.3}$$

The spectra are obtained by Fourier transforms, e.g.

$$I_{kl}(\omega) = \int_0^\infty (\cos \omega\tau)\phi_{kl}(\tau)d\tau \tag{5.4}$$

Each of the spectra is calculated in a similar fashion, so that we only consider $\phi_{xx}(\tau)$, which for simplicity we denote by $\phi(\tau)$.

If

$$\Phi_{xv}(\tau) = E\left\{\tilde{x}(t + \tau)\tilde{x}(t)\,\middle|\,\tilde{x}(t) = x, \tilde{v}(t) = v\right\} \tag{5.5}$$

is the conditional correlation function and $\rho(x,v,t)$ is the density for $(\tilde{x}, \tilde{v})$ at time t, then

$$\phi(\tau) = \iint \Phi_{xv}(\tau)\rho(x,v,t)dxdv \tag{5.6}$$

The ray method can be used to satisfactorily calculate $\Phi_{xv}(\tau)$, by integrating along rays that emanate from the point (x,v), (see I, §6 and Fig. 5).
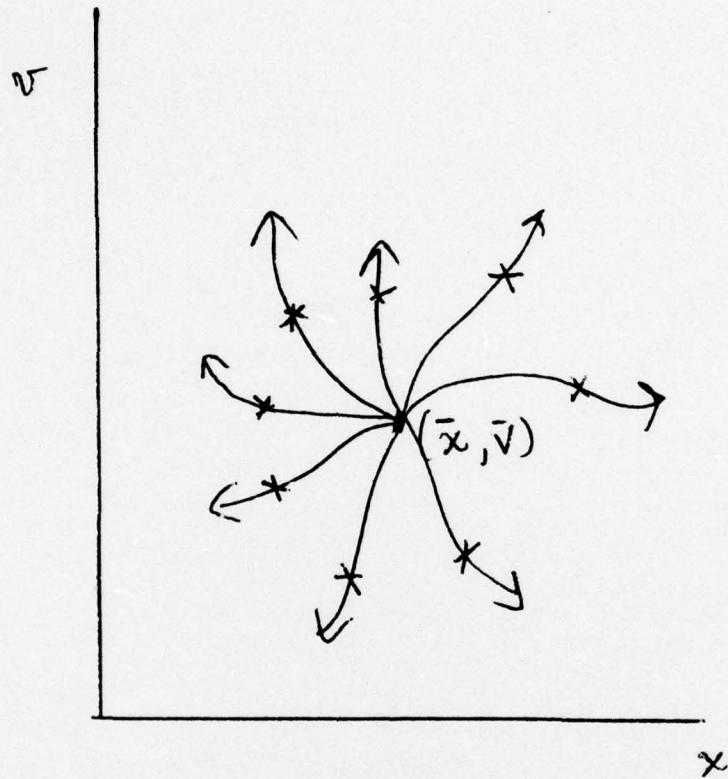
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FIG. 5: THE USE OF RAYS TO CALCULATE THE SPECTRUM. THE CROSS
MARKS INDICATE THOSE POINTS THAT CAN BE REACHED FROM
$(\bar{x}, \bar{v})$ IN A TIME τ BY FOLLOWING RAYS GENERATED FROM
(6.10). THE PROBABILITY DISTRIBUTION ON THESE RAYS IS
$\rho(\tau, x, v)$.

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SECTION 6. AN EXAMPLE

In this section we consider the dynamical equations

$$dx = vdt \qquad\qquad (6.1)$$

$$dv = (x^2 - \alpha - \gamma v)dt \qquad\qquad (6.2)$$

The potential function $V(x,\alpha) = -1/3x^3 + \alpha x$ is sketched in Fig. 6. In Figs. 7a-7e we sketch the (x,v) phase plane as α varies, for constant γ. As α decreases to zero, the height of the potential barrier $(Q = 4/3\alpha^{3/2})$ also decreases. In Fig. 8 we sketch contours of the transmission coefficient for $\varepsilon = .01$.
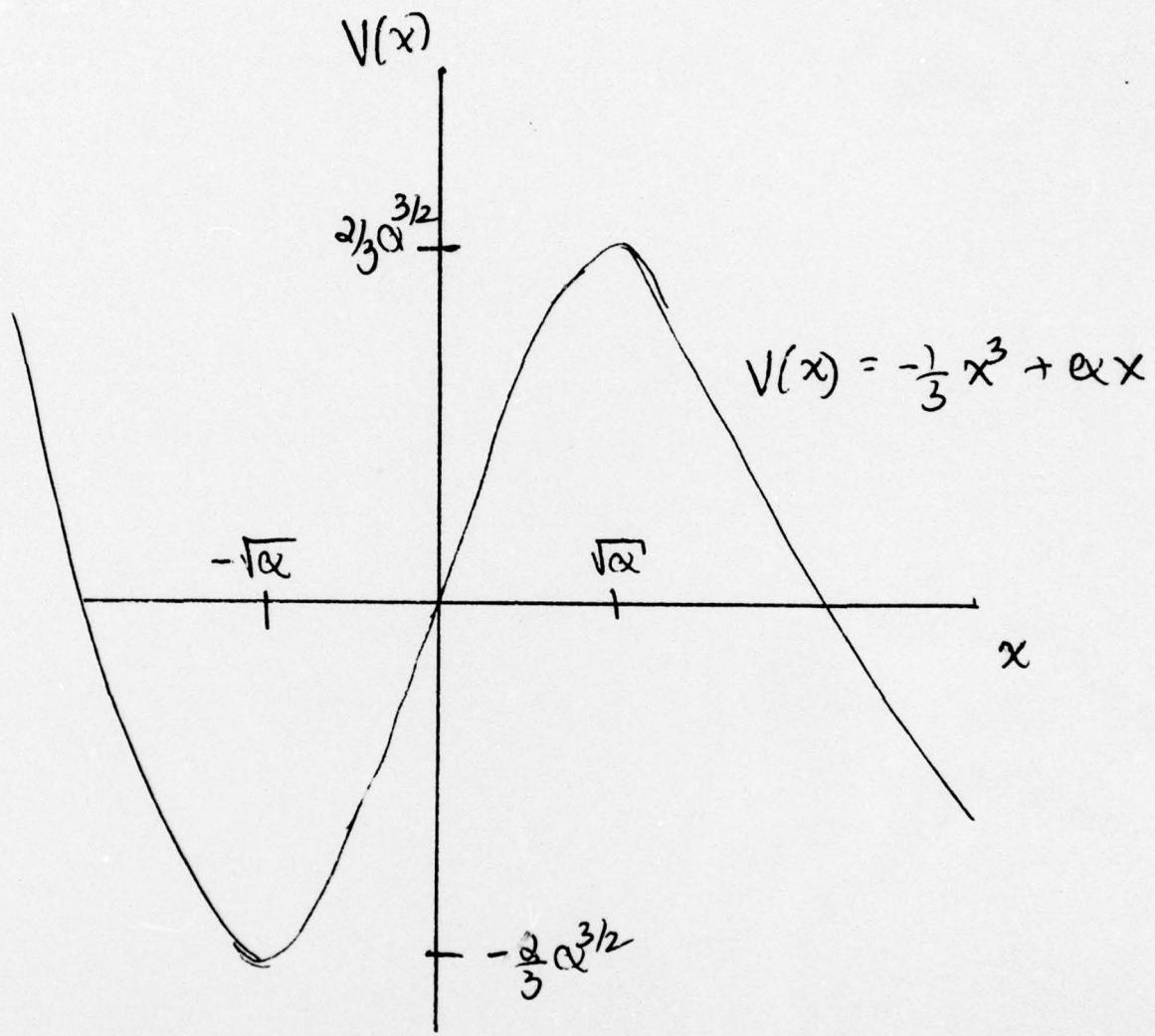
FIG. 6: POTENTIAL FUNCTION FOR THE SYSTEM CONSIDERED IN SECTION 6.

26
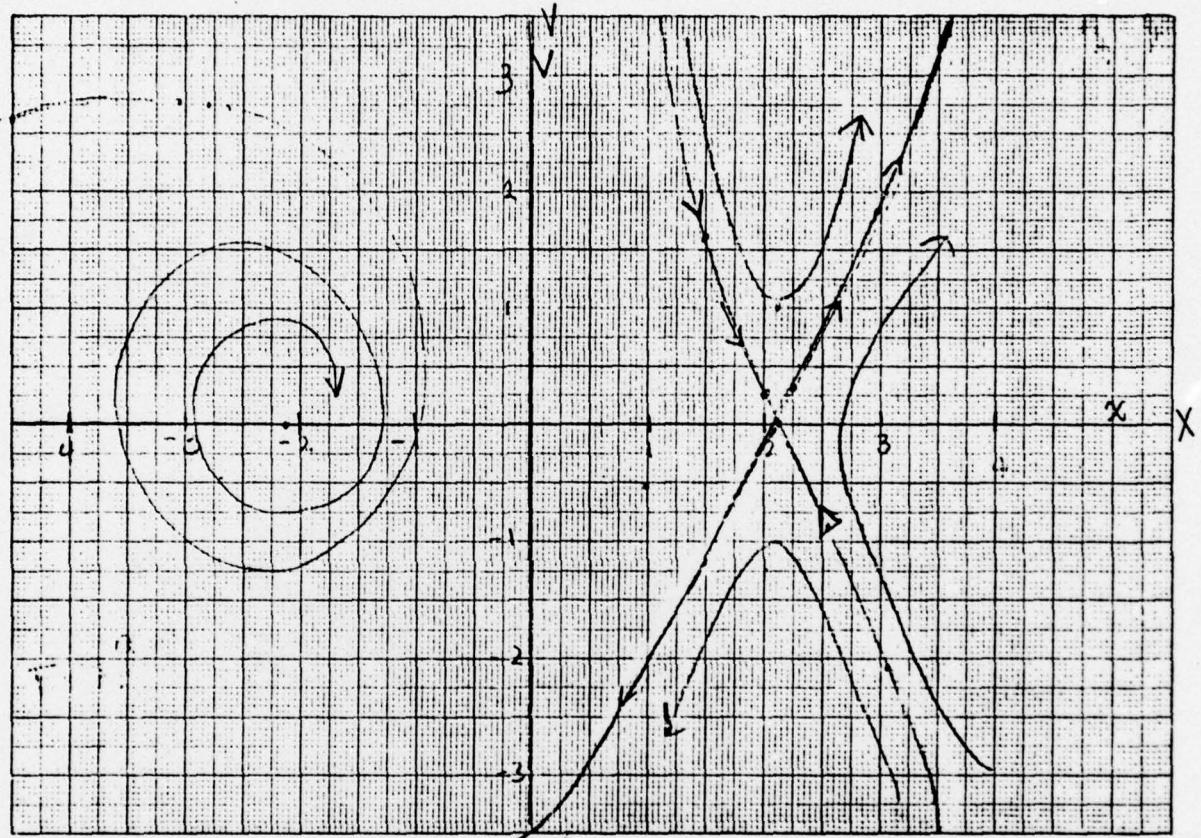
FIG. 7:   PHASE PLANE FOR THE DYNAMICAL SYSTEM

dx = vdt, dv = (x^2 - α - γv)dt.

A) α = 4.56, γ = .1

27
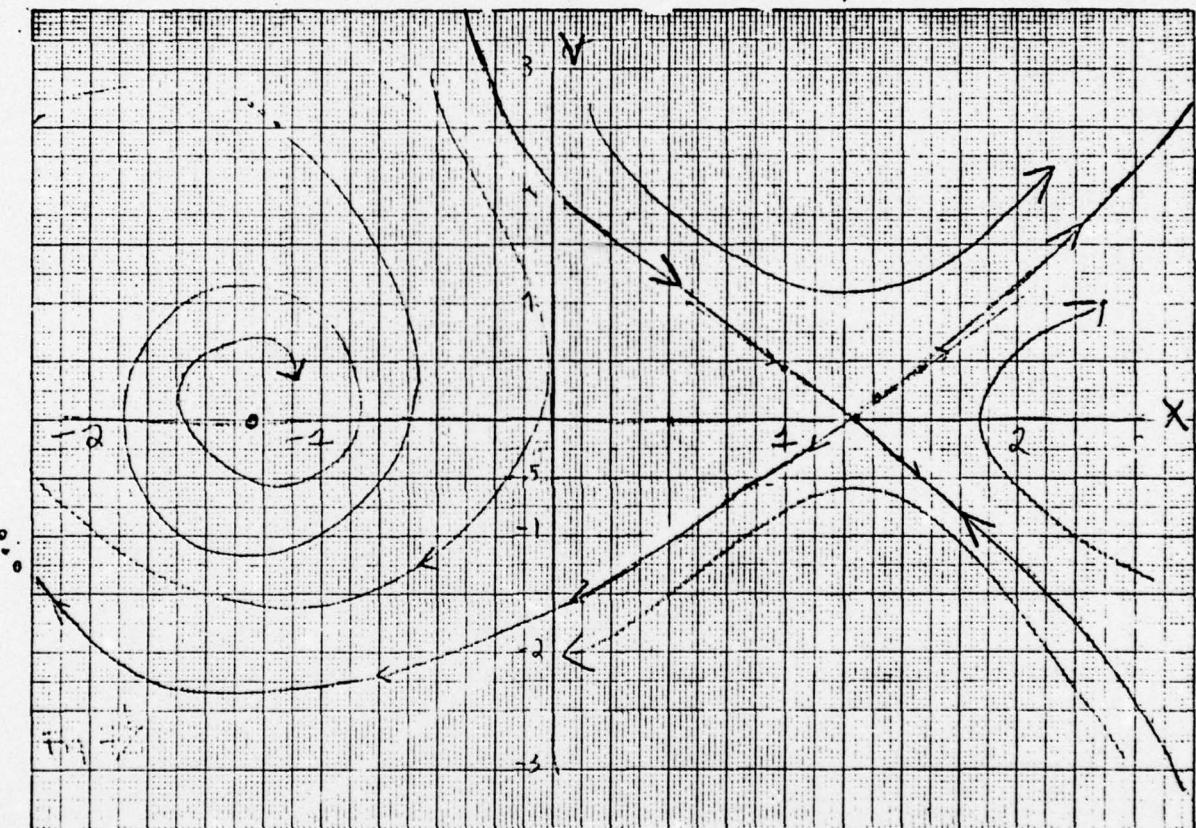
FIG. 7: (Continued)

B) $\alpha = 1.71$, $\gamma = .1$;

28
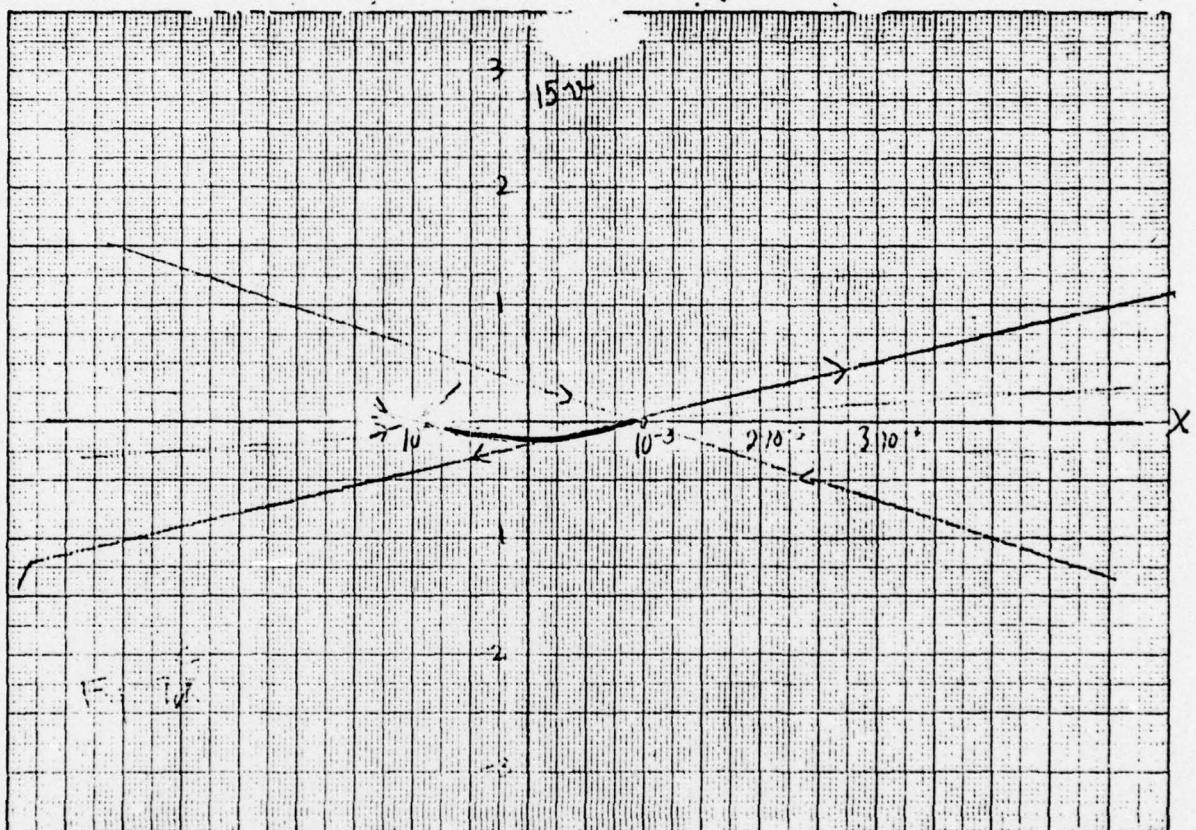
FIG. 7: (Continued)

C) $\alpha = 1.44 \times 10^{-6}$, $\gamma = .1$

29
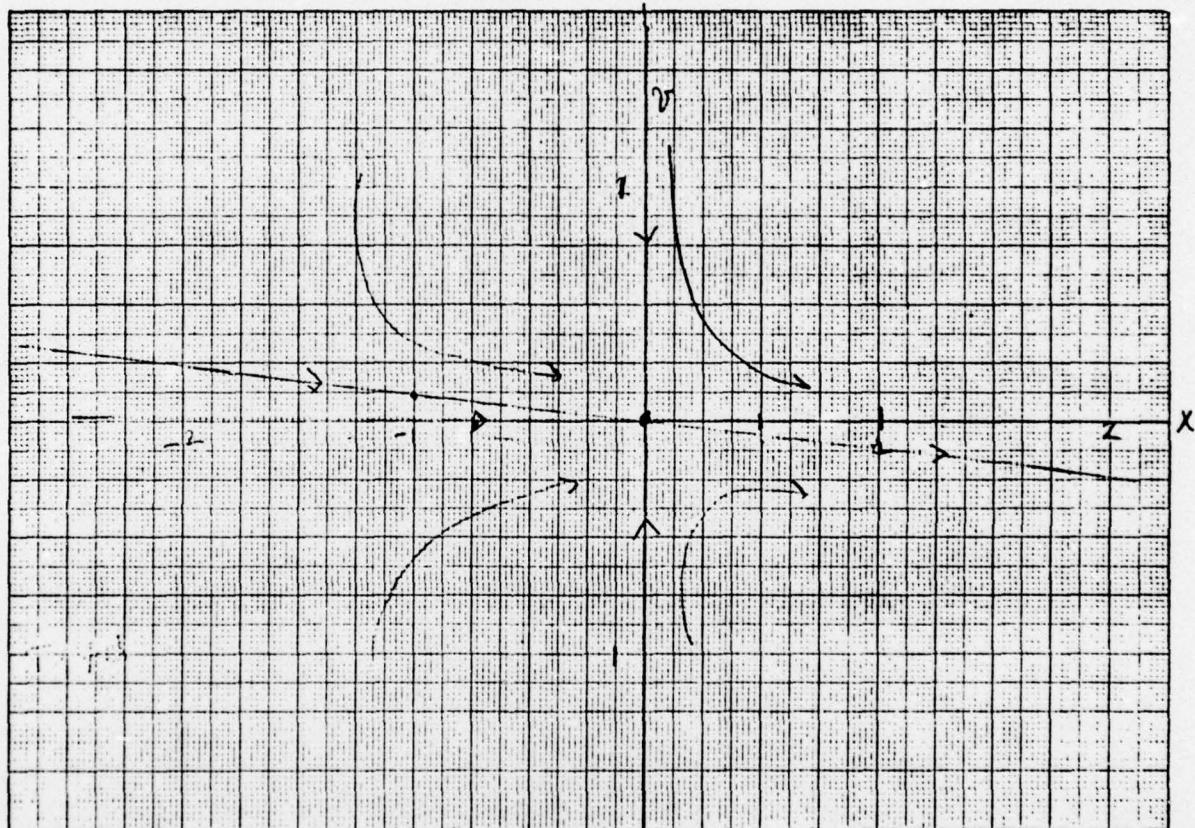
FIG. 7:   (Continued)

D) $\alpha = 0$, $\gamma = .1$

30
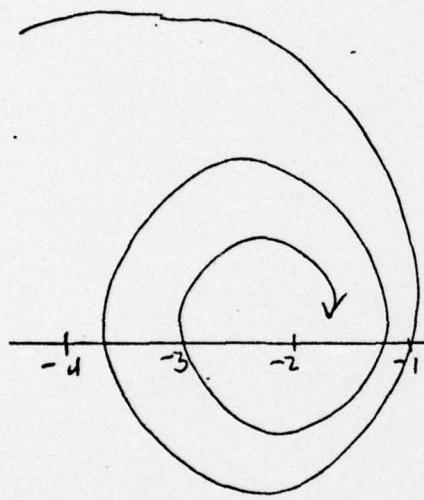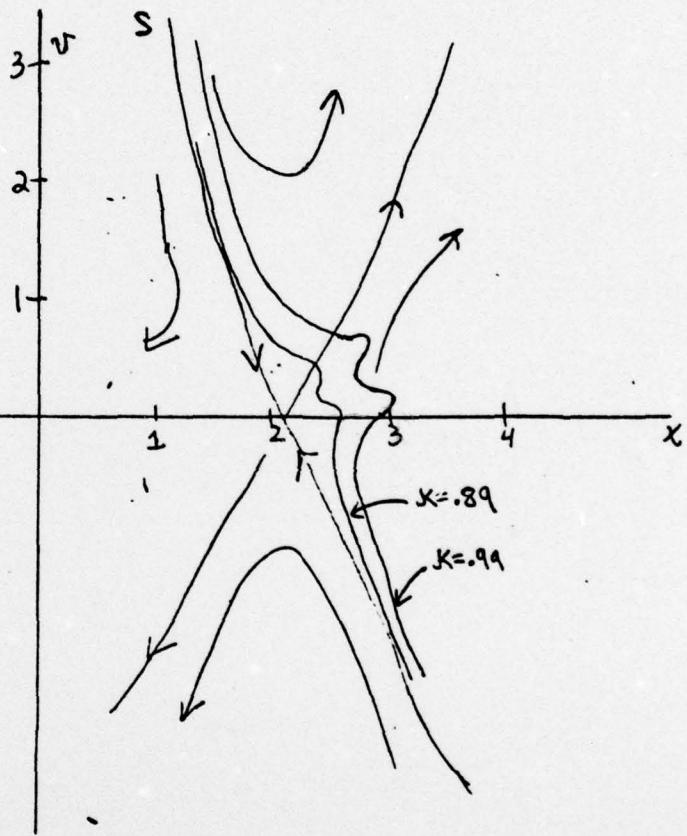
FIG. 8: CONTOURS FOR THE TRANSMISSION COEFFICIENT, FOR ϵ = .01.

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